Integrated Environmental Assessment and Management — Volume 1, Number 4—pp. 391–396

Actinides in Deer Tissues at the Rocky Flats Environmental Technology Site

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(Received 10 December 2004; Accepted 26 May 2005)

ABSTRACT

Limited hunting of deer at the future Rocky Flats National Wildlife Refuge has been proposed in U.S. Fish and Wildlife planning documents as a compatible wildlife-dependent public use. Historically, Rocky Flats site activities resulted in the contamination of surface environmental media with actinides, including isotopes of americium, plutonium, and uranium. In this study, measurements of actinides [Americium-241 (^{241}Am); Plutonium-238 (^{238}Pu); Plutonium-239,240 (^{239,240}Pu); uranium-233,244 (^{233,234}U); uranium-235,236 (^{235,236}U); and uranium-238 (^{238}U)] were completed on select liver, muscle, lung, bone, and kidney tissue samples harvested from resident Rocky Flats deer ($N = 26$) and control deer ($N = 1$). In total, only 17 of the more than 450 individual isotopic analyses conducted on Rocky Flats deer tissue samples measured actinide concentrations above method detection limits. Of these 17 detects, only 2 analyses, with analytical uncertainty values added, exceeded threshold values calculated around a 1×10^{-6} risk level (isotopic americium, 0.01 pCi/g; isotopic plutonium, 0.02 pCi/g; isotopic uranium, 0.2 pCi/g). Subsequent, conservative risk calculations suggest minimal human risk associated with ingestion of these edible deer tissues. The maximum calculated risk level in this study (4.73×10^{-6}) is at the low end of the U.S. Environmental Protection Agency's acceptable risk range.

Keywords: Actinides Tissue concentrations Refuge management Ungulates Human risk

INTRODUCTION

The Rocky Flats Environmental Technology Site (Rocky Flats), operated by the U.S. Department of Energy (USDOE), is a former nuclear weapons research, development, and production facility located northwest of Denver, Colorado, USA. Historical site activities included the fabrication of components for nuclear weapons from plutonium, uranium, beryllium, and stainless steel, and support activities included chemical recovery and purification of recyclable transuranic actinides. In 1992, the mission of the Rocky Flats site changed from weapons production to environmental cleanup and closure. Cleanup and remediation is being completed by the USDOE under oversight by the U.S. Environmental Protection Agency (USEPA) and the Colorado Department of Public Health and Environment.

By mandate of the Rocky Flats National Wildlife Refuge Act of 2001 [Pub. L. No. 107-107, 115 Stat. 102 (2001)] at site closure, portions of the site will become the Rocky Flats National Wildlife Refuge to be managed by the U.S. Fish and Wildlife Service (USFWS). Transfer of property is contingent on USEPA certification that cleanup and closure activities have been completed and that all monitoring and maintenance activities are operating properly and successfully.

The majority of the Rocky Flats site has remained undisturbed since its acquisition by the federal government and provides habitat for many wildlife species, including abundant populations of mule and white-tailed deer and seasonal populations of elk (Kaiser-Hill 2001). According to the National Wildlife Refuge System Improvement Act of 1997 [Pub. L. No. 105-57, 111 Stat. 1252 (1997)], the 6 wildlife-dependent priority public uses that must receive enhanced consideration in USFWS Refuge planning and management are hunting, fishing, wildlife observation, photography, interpretation, and environmental education. Future Rocky Flats Refuge lands will provide unique access for the disabled and youth to hunt in a controlled environment within close proximity to the Denver metropolitan area. Given that Rocky Flats ungulates have had access to actinide-contaminated areas (Symonds and Alldredge 1992), measurements of actinides in a range of tissues were needed to provide important information regarding potential human consumption risks and resultant compatibility of incorporating hunting as a recreational use on the Refuge.

The USFWS conducted this study to determine concentrations of selected actinides in relevant Rocky Flats ungulate tissues. Bone and kidney tissue samples were obtained because actinides are known to accumulate at higher concentrations in these organs (Whicker and Schultz 1982). Lung tissues were evaluated to assess actinide exposure to deer via the inhalation pathway. Finally, liver and muscle tissues were investigated because they are the tissues of the organism that are typically consumed by humans following a successful hunt. Analytical results from these edible tissues were used to carry out a series of conservative risk-based calculations to define human risk associated with ingesting these tissues.

STUDY AREA

The Rocky Flats Environmental Technology Site is a 6,240-acre property located approximately 16 miles northwest of Denver, Colorado, USA, and is bordered by Boulder, Broomfield, and Jefferson counties (Figure 1). Vegetation communities at Rocky Flats include unique xeric tallgrass prairie and tall upland shrubland, along with riparian woodland, riparian shrubland, wetlands, mesic mixed grassland, xeric needle and thread grassland, reclaimed mixed grassland and ponderosa pine woodland (USFWS 2004).

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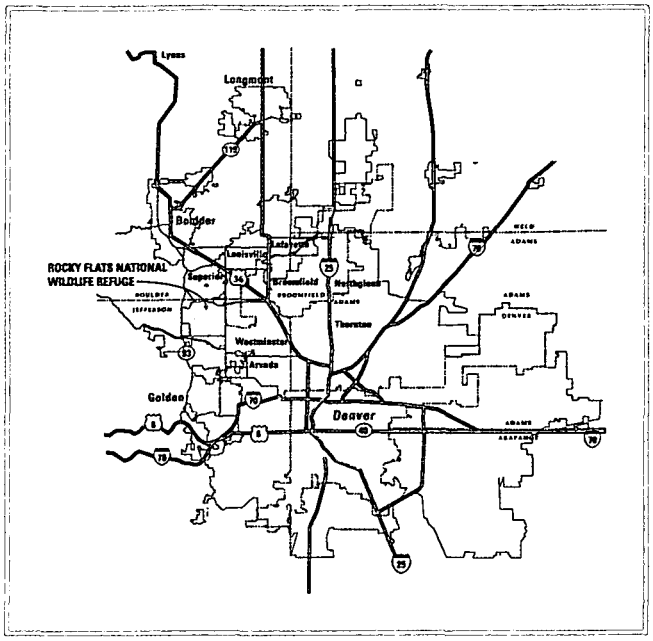

Figure 1. Location of the future Rocky Flats National Wildlife Refuge and surrounding communities.

METHODS

Field collection

Deer tissues were collected on the Rocky Flats site on 8 December 2002, during a chronic wasting disease study conducted by the Colorado Division of Wildlife. Twenty-six resident deer (24 mule, 1 whitetail, and 1 hybrid) were culled to test for chronic wasting disease and, at that time, USFWS biologists and 1 Rocky Flats ecologist harvested lung, liver, kidney, muscle, and bone tissues from the carcasses. Control tissue samples were obtained on 4 February 2004 from a mule deer killed by a vehicle at the Rocky Mountain Arsenal National Wildlife Refuge. Although the Rocky Mountain Arsenal was once a chemical weapons manufacturing center, a lack of historical radionuclide use on the site makes it an appropriate location for collection of reference deer tissues. The opportunistic harvest and subsequent analysis of tissues from this animal functioned, in conjunction with historical reference samples, as a qualitative indicator of regional, background radionuclide concentrations in deer tissues. It was decided that additional deer would not be culled for the expressed purpose of verifying low, "background" actinide concentrations. All tissues were rinsed with distilled water to remove any surface contamination and individually weighed, labeled, and double-bagged.

Bulk tissues remained frozen in a secure, sealed freezer (−10°C) at the Rocky Mountain Arsenal until 6 July 2004; at which point, subsamples were processed, packaged in ice, and shipped overnight to General Engineering Laboratories in Charleston, South Carolina, USA, for laboratory analyses. Although the normal contract sample-holding time for radiological samples is 180 d, the shortest half-life of the actinides of interest (^{241}Am, ^{238}Pu, ^{239,240}Pu, ^{233,234}U, ^{235,236}U, and ^{238}U) is 88 y (^{238}Pu). No appreciable loss of activity would have occurred during the 18 months between the time of collection and the time of isotopic analyses. Additional subsamples were resent on 21 July 2004 because of a laboratory error.

Actinide analyses

As the primary edible portions of the deer, all muscle and liver tissues were analyzed for all actinide isotopes of concern (^{241}Am, ^{238}Pu, ^{239,240}Pu, ^{233,234}U, ^{235,236}U, and ^{238}U). A subset of harvested lung, kidney, and bone tissues was analyzed for select actinides to obtain information regarding relative accumulation in nonedible tissues. In total, 90 tissue samples were analyzed for plutonium isotopes: 27 muscle, 27 liver, 6 kidney (composite), 15 lung, and 15 bone samples. Seventy-five sets of americium isotopic analyses were completed: 27 muscle, 27 liver, 6 kidney (composite), and 15 lung samples. Uranium analyses were conducted on 75 samples: 27 muscle, 27 liver, 6 kidney (composite), and 15 lung samples.

Analytical methodology was derived from a source method from the USDOE Environmental Measurements Laboratory Methods Manual and uses similar principles of radiochemical separation and counting (USDOE 1997). All samples were digested, if necessary, and aliquoted. Transuranic elements were scavenged by coprecipitation with iron hydroxide; the resultant precipitates were dissolved, and separation of elements was accomplished through the use of extraction chromatography and ion-exchange resins. Elements were then prepared for measurement of radioactive isotopes by coprecipitation with neodymium fluoride. Neodymium fluoride precipitates were trapped on filters, mounted on stainless steel disks, and placed in a partially evacuated chamber for the measurement of isotopic α emissions.

These analyses were performed according to General Engineering Laboratories method-specific quality control requirements, including proper instrument calibration and the use of method blanks, matrix spikes, sample duplicates, and tracer recovery.

Detection limits for analyses were needed that were lower than standard soil and water radiochemistry methods to detect actinide concentrations typical of tissue samples. To reach these levels, the laboratory used large sample sizes and longer count times. Calculations used to determine appropriate detection limits are presented below.

Calculation of reportable limits based on potential human risks

To ensure that detection limits were set sufficiently low to detect tissue concentrations of potential human concern via an ingestion pathway, the following calculations were carried out for each actinide isotope:

$$\text{Effective Dose Equivalent (Sv)} = \frac{\text{Risk Level}}{\text{Risk Coefficient (1/Sv)}} \quad (1)$$

$$\text{Radioactivity (Bq)} = = \frac{\text{Effective Dose Equivalent (Sv)}}{\text{Effective Dose Equivalent/Unit Intake (Sv} \cdot \text{Bq}^{-1})} \quad (2)$$

$$\text{Tissue Concentration (pCi} \cdot \text{g}^{-1}) = = \left[\frac{\text{Radioactivity (Bq)}}{\text{Edible Tissue Mass (kg)}}\right] \cdot \left(\frac{1\text{kg}}{1{,}000\text{g}}\right) \cdot \left(\frac{27\text{pCi}}{1\text{Bq}}\right) \quad (3)$$

Input values and resultant dose calculations are presented in Table 1.

Following these calculations, maximum detection limits required for analytical analyses were established as follows:

Table 1. Calculation of tissue actinide concentrations necessary for a 10^{-6} additional cancer risk, following human ingestion of all edible tissues from an average-sized Rocky Flats deer[ab]

Isotope	Risk level	Risk coefficient[c] (1/Sv)	EDE per unit intake[d] (Sv/Bq)	Edible tissue[e] (kg)	Dose calculation (pCi/g)	Detection limit (pCi/g)
^{239}Pu	1×10^{-6}	0.073	9.56×10^{-7}	30.3	0.0128	0.002
^{241}Am	1×10^{-6}	0.073	9.84×10^{-7}	30.3	0.0124	0.001
^{234}U	1×10^{-6}	0.073	7.66×10^{-8}	30.3	0.1594	0.02
^{238}U	1×10^{-6}	0.073	6.88×10^{-8}	30.3	0.1774	0.02

[a] Detection limits are set approximately an order of magnitude lower than this calculated value.
[b] EDE = effective dose equivalent; ^{239}Pu = plutonium-239; ^{241}Am = americium-241; ^{234}U = uranium-234; ^{238}U = uranium-238; Sv = sievert; Bq = becquerel; pCii = picocurie.
[c] The risk coefficient is needed to convert risk to effective dose. Value obtained from 1990 recommendations of the International Commission on Radiological Protection (ICRP 1992).
[d] The sum of the effective dose equivalents to various tissues of the body, each multiplied by its weighting factor. Effective dose equivalent per unit intake provides an estimate of the lifetime radiation dose to an individual from radioactive material taken into the body through either inhalation or ingestion (USEPA 1988). Values in the table are based on the most conservative, gastrointestinal absorption assumptions.
[e] An average deer was assumed to weigh 60 kg, of which, approximately 28 kg is edible muscle tissue and 2.3 kg is edible organ meat (liver).

isotopic americium, 0.001 pCi/g; isotopic plutonium, 0.002 pCi/g; and isotopic uranium, 0.02 pCi/g. These calculated values also serve as report thresholds (RT) for this study.

RESULTS

Contaminant analyses

In total, of the more than 450 individual isotopic analyses that were conducted on Rocky Flats deer tissue samples, 17 resulted in actinide concentrations measured above method detection limits (Table 2).

^{241}Am was detected in select lung, muscle, and kidney tissues of Rocky Flats deer and was also detected in kidney and liver tissues of the control deer from the Rocky Mountain Arsenal. Both measured isotopes of plutonium (^{238}Pu and ^{239,240}Pu) were detected only in select bone samples from Rocky Flats deer. Uranium isotopes (^{233,234}U, ^{235,236}U, and ^{238}U) were detected in select liver and muscle samples of Rocky Flats deer and were also detected in liver tissue of the control deer.

Radiological risk assessment

To predict potential radiological risk resulting from ingestion of edible deer tissues from Rocky Flats, a highly conservative estimate of risk was conducted. Uncertainty values were calculated as a function of counting efficiency error, peak area error, isotopic abundance error, systematic error, and sample calculated activity. Samples that yielded detectable quantities of any radioisotope were organized (Table 3), and analytical uncertainty values were added to measured results to produce a high-end estimate of tissue concentration. These values were compared with calculated report thresholds. Out of a total of 454 isotopic analyses, 2 yielded detectable concentrations of an actinide isotope of concern that, with the uncertainty value added, exceeded the precalculated report threshold (Table 3).

All liver and muscle tissues that yielded detects were used to back-calculate risk values associated with the ingestion of these tissues. As detailed above, uncertainty values were added to reported detection values to give a high-end estimate of tissue actinide concentrations. Bone, lung, and kidney tissues were not included in this analysis because they are not considered edible tissues for the purposes of this study. The same calculations that were used in the methods for setting detection limits were used in this analysis. A value of 2.3 kg was used as an approximate weight of edible organ meat (liver) from a 60-kg deer, and a value of 28 kg was used for the edible weight of muscle tissues.

On the basis of these conservative assumptions, the actinide risk associated with consuming the edible organ meats of an adult deer on Rocky Flats is presented in Table 3. The highest risk calculated in this exercise was attributable to ^{241}Am in the muscle tissue of deer 38-189-53, with tissue concentrations translating to a 6.76×10^{-8} risk level. This level of risk corresponds with a 1:14,700,000 increased chance of cancer resulting from ingestion of 28 kg of muscle tissue. If this same individual consumed this same amount of deer tissue yearly, throughout his or her lifetime (70 y), it would result in a 4.73×10^{-6} risk level, or a 1:210,000 increased chance of cancer. This risk level falls at the low-end of the USEPA's acceptable risk range of 1×10^{-4} to 1×10^{-6} (USEPA 1991).

DISCUSSION AND CONCLUSIONS

Historical investigations of actinide levels in Rocky Flats deer tissues yielded similar results (Table 4). In summary, plutonium analyses conducted on select tissues from 8 deer in the Rocky Flats region demonstrated tissue plutonium (^{238}Pu and ^{239,240}Pu) concentrations near or below detection limits (Hiatt 1977). Similarly, a total of 12 tissue samples (4 bone, 4 liver, and 4 lung) from Rocky Flats deer were analyzed for 2 plutonium isotopes in 1992, and all 24 analyses produced activities below detection limits (Symonds and Alldredge 1992).

The extremely low levels of actinides present in ungulate tissues at Rocky Flats are likely the result of low actinide levels across a majority of the site's surface soils, very low soil-to-plant actinide transfer rates (USEPA 1979; Hinton and Pinder 2001), and low gastrointestinal adsorption rates (ATSDR 1990), although actinide specific, terrestrial animal assimilation of actinides from the gastrointestinal tract is assumed to be less than 0.01% (Whicker and Schultz 1982).

Table 2. Comparison of measured concentrations of ^{241}Am, ^{238}Pu, $^{239,240}Pu$, $^{233,234}U$, $^{235,236}U$, and ^{238}U in lung, liver, muscle, bone, and kidney tissues of deer sampled at Rocky Flats and the Rocky Mountain Arsenal National Wildlife Reserve (control)[ab]

		^{241}Am		^{238}Pu		$^{239,240}Pu$		$^{233,234}U$		$^{235,236}U$		^{238}U	
Tissue	Statistic	RF	Control	RF	Control	RF	Control	RF	Control	RF	Control	RF	Control
Lung	Mean	0.000358	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
	N	2/14	0/1	0/14	0/1	0/14	0/1	0/14	0/1	0/14	0/1	0/14	0/1
	Max	0.000468	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Liver	Mean	bdl	0.000384	bdl	bdl	bdl	bdl	0.00673	bdl	0.00218	0.00192	bdl	bdl
	N	0/26	1/1	0/26	0/1	0/26	0/1	3/26	0/1	1/26	1/1	0/26	0/1
	Max	bdl	0.000384	bdl	bdl	bdl	bdl	0.0125	bdl	0.00218	0.00192	bdl	bdl
Muscle	Mean	0.000307	bdl	bdl	bdl	bdl	bdl	0.0033	bdl	bdl	bdl	0.00409	bdl
	N	4/26	0/1	0/26	0/1	0/26	0/1	1/26	0/1	0/26	0/1	1/26	0/1
	Max	0.000458	bdl	bdl	bdl	bdl	bdl	0.0033	bdl	bdl	bdl	0.00409	bdl
Bone	Mean	NA	NA	0.000623	bdl	0.000624	bdl	NA	NA	NA	NA	NA	NA
	N	NA	NA	2/14	0/1	2/14	0/1	NA	NA	NA	NA	NA	NA
	Max	NA	NA	0.000642	bdl	0.000773	bdl	NA	NA	NA	NA	NA	NA
Kidney	Mean	0.000983	0.000252	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
	N	1/5	1/1	0/5	0/1	0/5	0/1	0/4	0/1	0/4	0/1	0/4	0/1
	Max	0.000983	0.000252	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl

[a] ^{241}Am = americium-241; ^{238}Pu = plutonium-238; $^{239,240}Pu$ = plutonium-239,240; $^{233,234}U$ = uranium-233,234; $^{235,236}U$ = uranium-235,236; ^{238}U = uranium-238; Max = maximum; RF = Rocky Flats deer; *N* = number of detects/total number of samples analyzed; bdl = below detection level; NA = not applicable.

[b] Data are expressed as pCi/g wet tissue. The reported mean value is calculated as the arithmetic mean of all detects for a specific isotope within that tissue.

Table 3. Comparison of Rocky Flats deer tissue samples having actinides measurements above detection limits with calculated report thresholds[a]

Deer ID	Tissue type	Actinide isotope	Result (pCi/g)	Uncertainty (pCi/g)	Result + uncert. (pCi/g)	RT (pCi/g)	Above RT?	Calculated risk level
Rocky Flats tissues								
38-189-34	Bone	^{238}Pu	0.000603	0.000682	0.001285	0.002	No	—
38-189-39	Bone	^{238}Pu	0.000642	0.000629	0.001271	0.002	No	—
38-189-39	Bone	^{239,240}Pu	0.000475	0.000538	0.001013	0.002	No	—
38-189-44	Bone	^{239,240}Pu	0.000773	0.000678	0.001451	0.002	No	—
38-189-39	Lung	^{241}Am	0.000468	0.000375	0.000843	0.001	No	—
38-189-40	Lung	^{241}Am	0.000247	0.000280	0.000527	0.001	No	—
38-189-K5[b]	Kidney	^{241}Am	0.000983	0.000574	0.001557	0.001	Yes	—
38-189-31[c]	Liver	^{233,234}U	0.00302	0.00224	0.00526	0.020	No	2.51×10^{-9}
38-189-32[c]	Liver	^{233,234}U	0.00469	0.00261	0.0073	0.020	No	3.48×10^{-9}
38-189-52	Liver	^{233,234}U	0.0125	0.00926	0.02176	0.020	Yes	1.04×10^{-8}
38-189-54	Liver	^{235,236}U	0.00218	0.00191	0.00409	0.020	No	1.83×10^{-9}
38-189-48	Muscle	^{241}Am	0.000244	0.000276	0.00052	0.001	No	3.87×10^{-8}
38-189-49	Muscle	^{241}Am	0.000258	0.000292	0.00055	0.001	No	4.10×10^{-8}
38-189-50	Muscle	^{241}Am	0.000267	0.000302	0.000569	0.001	No	4.24×10^{-8}
38-189-53	Muscle	^{241}Am	0.000458	0.000449	0.000907	0.001	No	6.76×10^{-8}
38-189-41	Muscle	^{233,234}U	0.00330	0.00374	0.00704	0.020	No	4.08×10^{-8}
38-189-34	Muscle	^{238}U	0.00409	0.00400	0.00809	0.020	No	4.21×10^{-8}
Control tissue								
38-189-K6	Kidney	^{241}Am	0.000252	0.000285	0.000537	0.001	No	—
38-189-60	Liver	^{241}Am	0.000384	0.000435	0.000819	0.001	No	5.01×10^{-9}
38-189-60	Liver	^{235,236}U	0.00192	0.00188	0.0038	0.02	No	1.70×10^{-9}

[a] ^{241}Am = americium-241; ^{238}Pu = plutonium-238; ^{239,240}Pu = plutonium-239,240; ^{233,234}U = uranium-233,234; ^{235,236}U = uranium-235,236; ^{238}U = uranium-238; uncert. = analytical uncertainty values (uncertainty values were calculated as a function of counting efficiency error, peak area error, isotopic abundance error, systematic error, and sample calculated activity); RT = report threshold; pCi = picocurie.

[b] Kidney sample 38-189-K5 is a composite sample, with kidney samples from deer 38-189-44, 38-189-45, 38-189-48, 38-189-49, and 38-189-54.

[c] Tissues were involved in a laboratory fire during the ashing process; therefore, the reported values should be considered estimates of concentration. The other 2 samples involved in the fire (38-189-47 and 38-189-53) were nondetects.

The highly conservative risk levels (Table 3) likely overestimate risk associated with human ingestion of Rocky Flats deer tissues. A future hunting program at Rocky Flats National Wildlife Refuge would limit take of animals to a few individuals each year (USFWS 2004). Further, estimates of the mass of an average deer used in this study (60 kg) are higher than observed in comprehensive investigations of mule deer carcasses (42–52 kg) (Field et al. 2003). Finally, because analytical results are only slightly higher than detection limits, it is probable that several could be considered nondetects, given the equal magnitude of the uncertainty. Control samples in this study qualitatively support this possibility because several tissues from a site uncontaminated with actinide yielded detectable actinide concentrations comparable to samples from the Rocky Flats site (Table 2). Analysis of historical deer tissue control samples from Rocky Flats demonstrated similar results, with 2 of 9 tissue analyses indicating detectable concentrations of $Pu^{239,240}$ in deer from uncontaminated regions (Table 4).

As presented, this study functions as an indicator of maximum risk likely to result from human consumption of deer tissues from the Rocky Flats site under future management scenarios. Cleanup activities at Rocky Flats will continue until site closure, at which point all surface soils will meet stringent, human risk–based standards, as specified in the Final Rocky Flats Cleanup Agreement of 1996 (USEPA Region VIII, Colorado Department of Public Health and Environment, and USDOE federal facility agreement, signed 19 July 1996). As the cleanup at Rocky Flats concludes, levels of actinides in deer tissues may decrease, as contaminated surface soils will have been removed, resulting in fewer

Table 4. Historical measurements of actinide concentrations in Rocky Flats deer tissue samples[a]

Study	Tissue	Actinide	*N*	Max (pCi · g^{-1})
Hiatt 1977	Lung	^{238}Pu	1/7	0.0146
		$^{239,240}Pu$	5/7	0.0150
	Liver	^{238}Pu	0/6	bdl
		$^{239,240}Pu$	0/6	bdl
	Bone (metacarpal)	^{238}Pu	0/7	bdl
		$^{239,240}Pu$	1/7	0.0150
	Muscle	^{238}Pu	0/1	bdl
		$^{239,240}Pu$	0/1	bdl
	Control[b]	^{238}Pu	0/9	bdl
		$^{239,240}Pu$	2/9	0.0191 (liver)
Symonds 1992	Lung	^{238}Pu	0/4	bdl
		$^{239,240}Pu$	0/4	bdl
	Liver	^{238}Pu	0/4	bdl
		$^{239,240}Pu$	0/4	bdl
	Bone (rib)	^{238}Pu	0/4	bdl
		$^{239,240}Pu$	0/4	bdl

[a] ^{238}Pu = plutonium-238; $^{239,240}Pu$ = plutonium-239,240; Max = maximum; *N* = number of detects/total number of samples analyzed; bdl = below detection level; pCi = picocurie.

[b] Control tissues in this study were sampled from 1 lung, 3 livers, and 5 bones from 5 control deer outside of the Rocky Flats area. Detects occurred in the liver tissue sample of deer C1 and in the bone tissue sample of deer C4.

actinides available on the exterior surfaces of plants and/or assimilated into the plant itself.

Acknowledgement—Support for this work was provided through an interagency agreement (DE-AI34-02RFO2046) with the USDOE. We would like to thank Ward Whicker for his insight and comments on earlier drafts of this manuscript. We are also grateful to Michael Wedermyer and the Colorado Division of Wildlife for their support in the collection of tissue samples.

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